

Spin Gap of $S = 1/2$ Heisenberg Model on Distorted Diamond Chain

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Recently, Ishii et al.¹⁾ measured the magnetic susceptibility χ for $\text{Cu}_3\text{Cl}_6(\text{H}_2\text{O})_2 \cdot 2\text{H}_8\text{C}_4\text{SO}_2$, which is considered to be a quasi-one-dimensional material consisting of $S = \frac{1}{2}$ trimer spin chains. The result indicates that χ vanishes in the low temperature limit. They also measured the magnetization process for this material, and showed that there is a plateau of zero magnetization below the critical field $H_c \simeq 3.9$ T. From these experimental results, they concluded that the ground state is a singlet state with spin gap. The spin gap Δ is estimated as $\Delta/k_B \simeq 5.2$ K from the value of H_c .

The proposed Hamiltonian¹⁾ representing a spin chain in this material is given by

$$H = J_1 \sum_j (\mathbf{S}_{3j-1} \cdot \mathbf{S}_{3j} + \mathbf{S}_{3j} \cdot \mathbf{S}_{3j+1}) + J_2 \sum_j \mathbf{S}_{3j+1} \cdot \mathbf{S}_{3j+2} + J_3 \sum_j (\mathbf{S}_{3j-2} \cdot \mathbf{S}_{3j} + \mathbf{S}_{3j} \cdot \mathbf{S}_{3j+2}), \quad (1)$$

where \mathbf{S}_j is the $S = \frac{1}{2}$ spin on site j . Three spins \mathbf{S}_{3j-1} , \mathbf{S}_{3j} and \mathbf{S}_{3j+1} form a trimer. The lattice structure is shown in Fig. 1. Three kinds of exchange constants J_1 , J_2 and J_3 are inferred to be positive and to satisfy the relation $J_1 > J_2, J_3$ from the lattice parameters of the material.²⁾ Hereafter, we use the unit of $J_1 = 1$.

The symmetric case of $J_3 = J_1 (= 1)$ has been studied by Takano et al.³⁾ and the system has been called the *diamond chain*. They almost exactly showed that there exist three phases in the parameter space; the ferrimagnetic

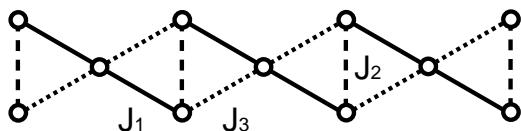


Fig. 1. The lattice structure of the distorted diamond chain representing $\text{Cu}_3\text{Cl}_6(\text{H}_2\text{O})_2 \cdot 2\text{H}_8\text{C}_4\text{SO}_2$. Three kinds of exchange constants are shown.

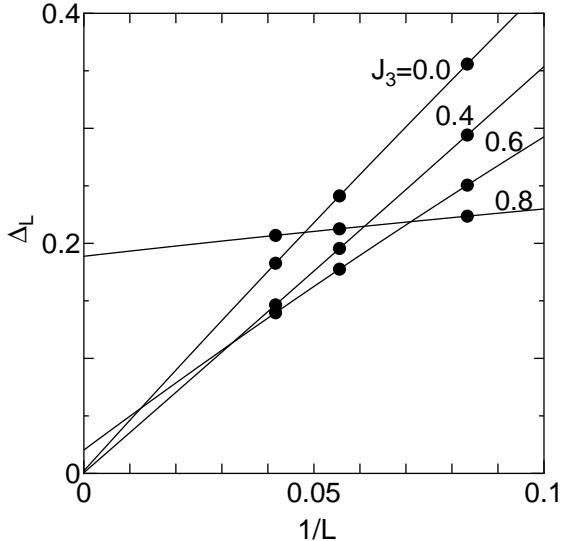


Fig. 2. Spin gaps of finite systems with $L = 12, 18$ and 24 (solid circles) for several values of J_3 at $J_2 = 1$. Each solid line represents eq. (2) with fitting parameters c_1 , c_2 and Δ_∞ for a value of J_3 .

phase for $J_2 < 0.909$, the tetramer-dimer (TD) singlet phase for $0.909 < J_2 < 2$ and the dimer-monomer (DM) singlet phase for $J_2 > 2$. The TD phase is a disordered phase with spin gap which originates from frustration among exchange interactions, while the DM phase is a spin fluid phase without spin gap. Okamoto et al.⁴⁾ studied the general case of $J_3 \neq 1$; i. e. the *distorted diamond chain*. The three phases develop in the J_2 - J_3 plane. They numerically determined the phase boundaries. Also Tonegawa et al.⁵⁾ numerically studied the magnetization process and showed plateaux for $\frac{1}{3}$ and $\frac{2}{3}$ of the saturation field.

In this article, we estimate the values of the spin gap by the numerical diagonalization. Then we produce a contour map in the J_2 - J_3 parameter space. The contour map represents an overall feature of the gapped phase of the $S = \frac{1}{2}$ Heisenberg model on the distorted diamond chain. When further experimental information on $\text{Cu}_3\text{Cl}_6(\text{H}_2\text{O})_2 \cdot 2\text{H}_8\text{C}_4\text{SO}_2$ is given, the contour map will be useful to determine the values of the exchange constants for the real material.

We first calculate the spin gap Δ_L for finite chains with system size L . The spin gap Δ_∞ in the thermodynamic limit is evaluated by extrapolation. We assume the size dependence of Δ_L as

$$\Delta_L = \Delta_\infty + \frac{c_1}{L} + \frac{c_2}{L^2} \quad (2)$$

with constants c_1 and c_2 . The numerical diagonalization has been done for $L = 12, 18$ and 24 under the periodic boundary condition. We determine c_1 , c_2 and Δ_∞ by fitting. In Fig. 2, we show Δ_L as a function of L for several values of J_3 at $J_2 = 1$. For $J_3 = 0$, the estimated value of Δ_∞ is about 0.002 and is close to zero; the nonzero value is interpreted as an extrapolation error.⁶⁾ For $0 < J_3 \lesssim 0.4$, the true value of the spin gap is very small or may be regarded as zero, since the estimated

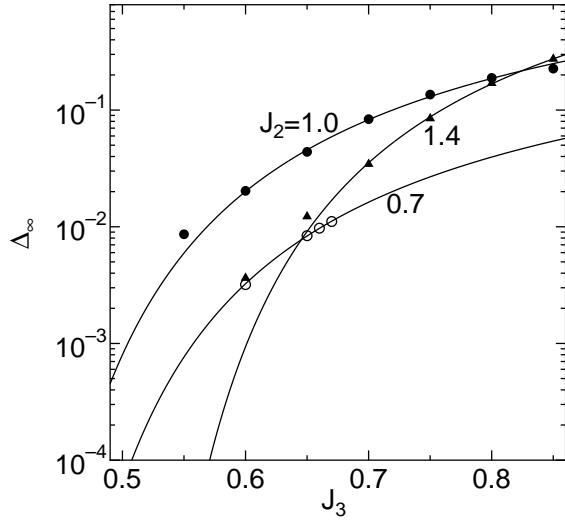


Fig. 3. The spin gap Δ_∞ in the thermodynamic limit (symbols) as a function of J_3 for $J_2 = 0.7, 1.0$ and 1.4 . Each solid line represents $D(J_3)$ (eq. (3)) with fitting parameters a_1 and a_2 for a value of J_2 .

values are less than 0.002 and are within the extrapolation error. For $J_3 \gtrsim 0.6$, the figure shows that the system has a finite spin gap. For $J_3 = 0.5$, Δ_∞ is 0.0034, which is small but seems to be finite. This is consistent with the result of Okamoto et al. that the spin gap opens at the critical value $J_3^c \simeq 0.35$ for $J_2 = 1$.⁴⁾

In general, the spin gap in a dimer phase is exponentially small near the phase boundary to a spin fluid phase. Hence it is difficult to estimate Δ_∞ near the boundary in the present case. To overcome this difficulty, we assume that J_3 dependence of the spin gap is given by

$$D(J_3) = a_1 \sqrt{J_3 - J_3^c} \exp\left(-\frac{a_2}{J_3 - J_3^c}\right) \quad (3)$$

for $J_3 \sim J_3^c$,⁷⁾ where a_1 and a_2 are constants. We have the values of J_3^c by inspecting the phase diagram of Okamoto et al.;⁴⁾ e.g. $J_3^c = 0.374, 0.354$ and 0.460 for $J_2 = 0.7, 1.0$ and 1.4 , respectively. We carry out the fitting of the extrapolation data Δ_∞ to eq. (3) and determine a_1 and a_2 . Figure 3 represents the fitting function $D(J_3)$ and the extrapolation data. We find that the extrapolation data are well reproduced by eq. (3) for $\Delta_\infty \gtrsim 0.02$. Hence the function form in eq. (3) is reliable. We use eq. (3) to estimate the spin gap for $\Delta_\infty \lesssim 0.02$ near the critical value J_3^c . For example, the spin gap is estimated as 1.0×10^{-2} at $J_3 \simeq 0.57$, 1.0×10^{-3} at $J_3 \simeq 0.50$, 1.0×10^{-4} at $J_3 \simeq 0.47$ and 1.0×10^{-5} at $J_3 \simeq 0.45$ for $J_2 = 1.0$.

Using these results, we draw contour lines of the spin gap in the J_2 - J_3 plain. The resultant contour map is shown in Fig. 4. We have calculated Δ_∞ at the discrete positions (J_2, J_3) with $J_2 = 0.7, 0.8, \dots, 2.0$ and $J_3 = 0.5, 0.55, \dots, 1.0$. For $\Delta_\infty > 0.02$, the positions of solid circles are determined by the linear interpolation among the spin gaps Δ_∞ at the discrete positions. For $\Delta_\infty < 0.02$, the positions of open circles are determined

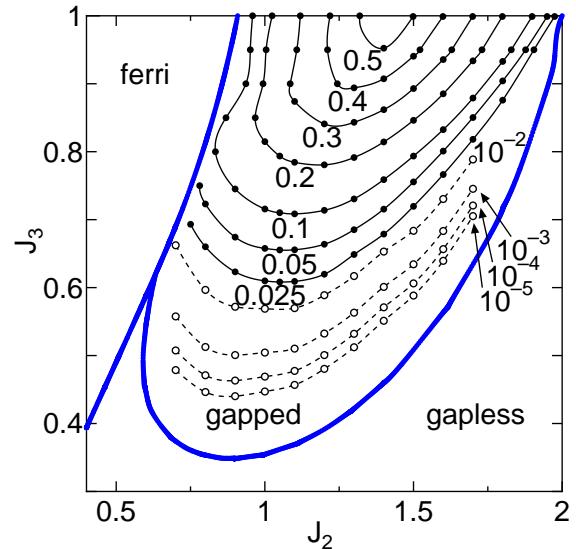


Fig. 4. The contour map for the spin gap in the gapped phase of the J_2 - J_3 plane. Bold solid lines are the phase boundaries that Okamoto et al. have determined.⁴⁾

by using $D(J_3)$ (eq. (3)) instead of Δ_∞ .

The temperature dependence of the experimental magnetic susceptibility has a broad peak at ~ 70 K. It suggests that the energy scale of the characteristic exchange constant J_1 is larger than 70 K. Here we consider a case of J_1 being 100 K as an example.⁸⁾ In this case, we have $\Delta_\infty \sim 0.05$ according to the observed spin gap ~ 5 K. Then J_2 and J_3 are limited to values close to the contour line of $\Delta_\infty = 0.05$ and of $J_2 < 1$.

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